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Effects of antioxidant on mechanical and electromagnetic properties of carbonyl iron/ethylene propylene diene monomer composites

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ABSTRACT

In this paper, we study the effect of the loading of poly(1,2-dihydro-2,2,4-tri-methyl-quinoline) (antioxidant RD) on the carbonyl iron powder (CIP)/ethylene propylene diene monomer (EPDM) composite absorbing material under high-temperature environment.Through macro- and micro-analysis, it is found that the composite material loaded with 0 phr antioxidant RD fail due to the aging of the rubber matrix at 150 °C for 2 days.When the load is 3 phr, the service life exceeds 10 days.In the electromagnetic performance test, the minimum reflectance of the sample with the antioxidant RD load of 0 phr increased from − 22.16 to − 17.39 dB within 2 days, and the effective absorption bandwidth (RL < − 10dB) was reduced from 2.23 to 2.04 GHz.And the minimum reflectance of the sample with the antioxidant RD loading of 3 phr increased from − 19.91 to − 16.81 dB within 10 days, and the effective absorption bandwidth was reduced from 2.25 to 2.06 GHz with small fluctuations.Therefore,loading antioxidant RD can effectively delay the aging rate of CIP/EPDM composite absorbing materials.

1 Introduction

Microwave absorbing materials mainly absorb electromagnetic waves in specific frequency bands in electronic instruments to ensure the stable operation of the equipment [1–3].Microwave absorbing materials are often faced with harsh working environments such as high vacuum [4–6], high frequency, and high power and need to be adapted to device cavities with complex structures [7, 8]. This requires the material to have strong attenuation [9, 10], good heat resistance, and should be easy to process and shape [11].

The rubber-based composite absorbing material has become a potential absorbing material because of its easy processing and shaping, softness, good adhesion, and strong attenuation [12, 13]. EPDM, because its main chain is composed of saturated hydrocarbons, contains unsaturated bonds only in the side chain, so it has excellent resistance to ozone, heat, climate and other aging performance, and excellent electrical insulation performance and has

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been widely concerned [14, 15]. Katiyar et al. [16] prepared a manganese-zinc ferrite/EPDM composite absorbing material and found that at a frequency of 4GHz, the real part of the complex permittivity increased from 2.4 to 8.1, the imaginary part increased from 0.06 to 0.55, the real part of the complex permeability increased from 1.0 to 1.48, and the imaginary part increases from 0 to 1.3. Wang et al. [17] prepared CNTs/EPDM composite absorbing materials and found that CNTs/EPDM composite absorbing materials have good absorbing performance in the 5-18 GHz bandwidth; when the content of CNTs exceeds 40%, the lowest reflectivity starts to increase, the absorbing performance decreases, and the material will be reflective.

Carbonyl iron powder has the advantages of high Curie temperature, high thermal stability, and high microwave permeability, so it is widely used as an absorbent in absorbing materials [18–20]. Feng et al. [21] prepared CIP/EPDM composite absorbing materials by filling carbonyl iron powder as an absorbent in EPDM and studied the influence of CIP content on its electromagnetic and absorbing properties. It is found that when the CIP volume fraction is 45%, the lowest reflectivity of the material reaches − 21.7 dB at 3.5 GHz.

To date, there have been many researches on rubber-based composite absorbing materials [22–24], but there are few reports on improving the aging resistance of rubber-based composite absorbing materials [25–27]. Thermal aging of rubber belongs to free radical chain autocatalytic oxidation reaction. Assink et al. [28] monitored the thermal degradation of EPDM by hydrogen nuclear magnetic resonance spectroscopy and found that the scission of macromolecular chains was the main thermal behavior at the initial stage of thermal degradation, and the oligomers caused by chain scission in the later stage of thermal degradation were mainly cross-linked. Pi et al. [29] found that the surface of EPDM samples became rough with aging time, cracks appeared, and the original elasticity was lost.

Antioxidant RD is a kind of ketamine anti-aging agent. It is used as a free radical inhibitor to stop the chain reaction from the free radical part by hydrogen atom donation, free radical and alkyl radical capture, and electron donation, which can prevent rubber aging caused by high temperature. It has an excellent protective effect and also has a strong inhibitory effect on the catalytic oxidation of metals [30]. Hu et al. [31] found that when the weight ratio of RD to MB is 0.5, the TE index of EPDM after vulcanization at 175 °C and 24 h is above 65.

Herein, we have prepared a 45% volume fraction of CIP-filled EPDM to make composite absorbent materials. The electromagnetic and mechanical properties of EPDM/CIP composite absorbing materials loaded with different amounts of antioxidant RD under 150 °C hot air aging conditions were studied.

2 Experiment part

2.1 Material

EPDM (4570; Vinyl content = 50%; ENB content = 4.9%) was purchased from Dow Chemical Company.Carbonyl iron powder (YW1; D_{50} = 3.5 μm, the particle size distribution is shown in Fig. 1; Fe content > 98%) was purchased from Jiangsu Tianyi Ultra-fine Metal Powder Co. LTD. Silane coupling agent (KH550; Purity ≥ 98%) was purchased from Nanjing Chuangshi Chemical Co. LTD.Vulcanization agent (2,5-Dimethyl-2 ,5-di (tert-butylperoxy) hexane; DPBMH; Purity ≥ 93%) was purchased from Shanghai Sendi Chemical Co. LTD. Antioxidant RD (poly(1,2-dihydro-2,2,4-trimethyl-quinoline); Purity ≥ 98%) was purchased from Guangdong Duba New Material Technology Co. LTD.

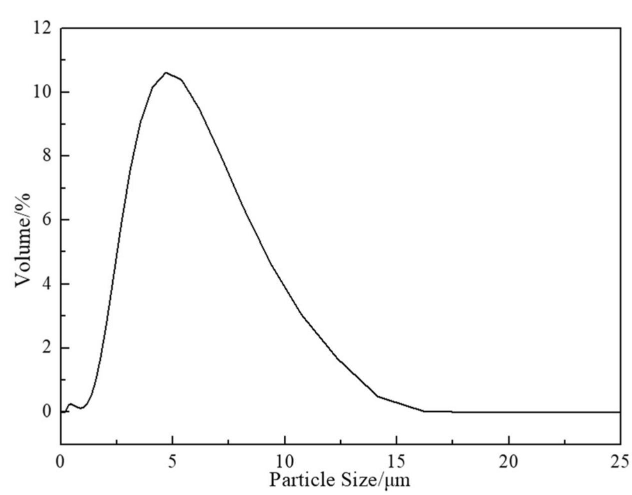


Fig. 1 Particle Size Distribution

2.2 Sample preparation

The compounding formulation is shown in Table 1 .The specific preparation process of the CIP/EPDM composites is shown in Fig. 2. CIP and silane coupling agent KH550 with a mass ratio of 100:2 were mixed evenly in the mixer.Then a certain amount of EPDM (EPDM:CIP = 55:45, volume ratio) was placed in the mixing machine for mixing. After the EPDM was completely coated, the weighed antioxidant RD, the CIP after KH550 treatment, and the 2,5-Bis(tert-butylperoxy)-2,5-dimethylhexane (the dosage was 3 wt% of the mass of EPDM) were added to the mixing machine for mixing with EPDM successively to obtain the mixing rubber.A certain amount of evenly mixed CIP/EPDM mixture was vulcanized by a plate vulcanizer to obtain a CIP/EPDM composite absorbing material vulcanization film for aging behavior research.A certain amount of evenly mixed CIP/EPDM mixture was vulcanized in a ring mold to form a coaxial ring with a thickness of 2.8 mm, an inner diameter of 3.04 mm, and an outer diameter of 7 mm, which was used to study the wave absorption and electromagnetic properties of CIP/EPDM composite absorbing materials.

2.3 Material characterization and measurement

The mechanical properties were tested by using an electromechanical universal testing machine (CMT 5254, shenzhen SANS testing machine, China). The ATR-FTIR spectrum of EPDM/CIP composite absorption material samples before and after hot air aging was determined by Nicolet spectrometer (nexus670 technology), with a resolution of 4 cm^{-1} and a range of 400–4000 cm^{-1}. The thermogravimeter of Netzsch STA449F1 was used for the linear analysis between 25 °C and 1000 °C, with the heating rate of 20 °C min^{-1} and the air flow rate of 30 mL min^{-1}. Each sample was measured in an alumina crucible weighing 5–10 mg. The scanning electron microscope (SEM) observation was carried out by Hitachi TM3000 scanning electron microscope under the condition of 15 kv acceleration voltage.

3 Result and discussion

3.1 Influence of different contents of RD on mechanical properties of CIP/EPDM composite absorbing materials before and after hot air aging

Changes in the mechanical properties of materials reflect the length of service life of materials from a macro perspective. Figure 3 shows the changes of mechanical properties of CIP/EPDM composite absorbing materials with different contents of RD before and after hot air aging. It can be seen from the figure that the aging trend of CIP/EPDM composite absorbing materials has slowed down obviously with the increase of RD.When RD content was 0, the composites lost mechanical properties after 2 days of aging.When RD content was 1.0 phr, the composite absorbing material lost its mechanical properties after 5 days of aging.When RD content increased to 1.5 phr, the failure time was extended to about 8 days. When RD content is greater than 2.0 phr, the material still has certain mechanical properties after 10 days.Compared with CIP/EPDM composite absorbing materials without anti-aging agent, they lose their mechanical properties after aging for over 48 h. This indicates that the antioxidant RD delays the molecular chain breakage and cross-linking process of rubber samples. The anti-aging effect of antioxidant RD is obvious, and the more the antioxidant RD loaded, the better the anti-aging effect is. However, in

Table 1 Compounding formulation (phr)

Materials	Purpose	Loaded 0 phr RD	Loaded 1 phr RD	Loaded 1.5 phr RD	Loaded 2 phr RD	Loaded 3 phr RD
EPDM	Basis material	100	100	100	100	100
CIP	Absorbing agent	720	720	720	720	720
KH550	Coupling agent	14.4	14.4	14.4	14.4	14.4
RD	Antioxidant	–	1	1.5	2	3
DPBMH	Vulcanizing agent	3	3	3	3	3

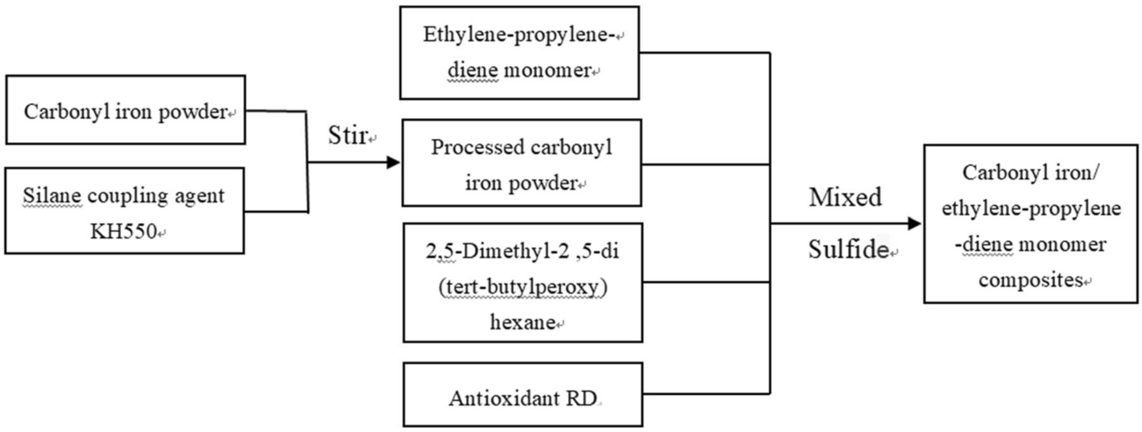


Fig. 2 Experimental flow chart

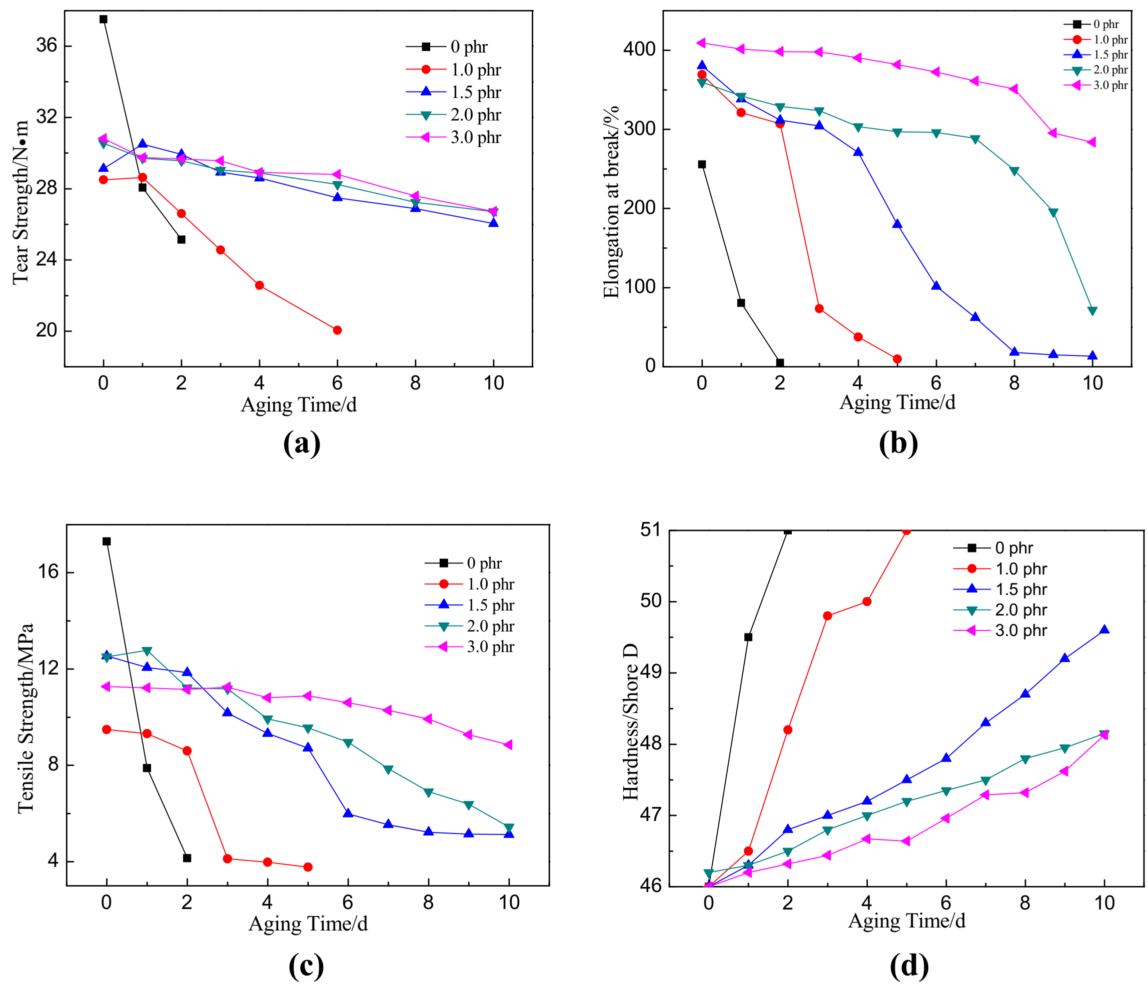


Fig. 3 Changes in mechanisms properties of CIP/EPDM composite absorbing material with different loading fractions of antioxidant RD: **a** tear strength; **b** elongation at break; **c** tensile strength; and **d** shore D hardness

the actual operation process, it was found that when the antioxidant loading exceeds 3.0 phr, the rubber will bloom and white crystals appear on the rubber surface, so the loading of antioxidant RD should not exceed 3.0 phr, and the best loading is 2.0 to 3.0 phr.

3.2 SEM before and after the hot air aging of CIP/EPDM composite absorbing material with different dosage of antioxidant RD

In order to prove that the antioxidant RD has the effect of delaying the aging phenomenon of CIP/ EPDM composite materials, SEM analysis was carried out. Figures 4, 5, and 6 are SEM images of CIP/ EPDM composite absorbing materials with 0 phr, 1.0 phr, and 3.0 phr antioxidant RD before and after hot air aging, respectively. From Fig. 4c in the aging after 16 h, material surface becomes rough, and CIP particles are part exposed to the material surface; this is because the composite absorbing material has more oxygen on its surface as it ages, the surface oxygen content is more, the surface of EPDM oligomer produced by the oxidation degradation under the action of CIP and oxygen, oxidizing and cross-linking reaction, and cross-linking of small molecules escape from the surface under the action of a heat flow. From Fig. 5d, it can be seen that by adding 1.0 phr antioxidant RD,after 8 days of aging , rough surface and gap increased, and CIP particles from the phenomenon of EPDM continuous phase were exposed. It can be seen from Fig. 6 that loading 3.0 phr antioxidant RD, after 14 d hot air aging, the composite absorbing material's surface still keep a good shape, and CIP evenly dispersed in continuous phase.The above results show that adding antioxidant RD can effectively inhibit the occurrence of cross-linking reaction between CIP and O_2 in the hot air aging process.The results further indicate that the antioxidant RD can effectively inhibit the hot air aging of CIP/EPDM composite materials.

3.3 ATR-FTIR before and after hot air aging of CIP/EPDM composite absorbing material with different doses of antioxidant RD

The ATR-FTIR analysis of samples with different antioxidant RD content proved that antioxidant RD has a delaying effect on the aging phenomenon of CIP/EPDM composites from a microscopic point of view. Figure 7 shows absorbance of CIP/EPDM composite material with different dosage of antioxidant RD at different time of hot air aging. The absorption peaks at 2920 cm^{-1} and 2851 cm^{-1} correspond to the asymmetric stretching vibration peaks of aliphatic alkanes on the saturated C–H framework of EPDM. The characteristic peaks at 1465 cm^{-1} and 1375 cm^{-1} are the shear vibration peaks of $-CH_2-$ on the typical EPDM and the C–H symmetric stretching vibration peaks of the propylene part $-CH_3$. The peak at 1685 cm^{-1} corresponds to the C=C stretching

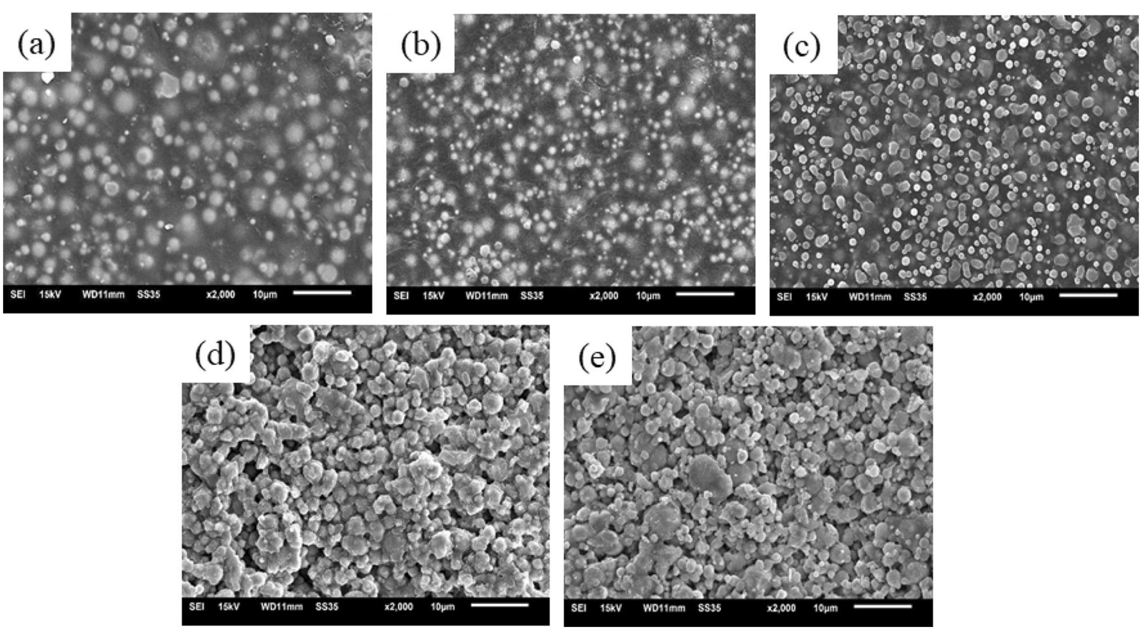


Fig. 4 SEM images of the surface of CIP/EPDM composite absorbing material with 0 phr of antioxidant RD before and after hot air aging: **a** 0 h, **b** 8 h, **c** 16 h, **d** 24 h, and **e** 48 h

vibration absorption peak. The characteristic peak at 718 cm^{-1} corresponds to the continuous methylene vibrational peak on the EPDM framework. It can be clearly seen from the figure that with the increase of

Fig. 7 ATR-FTIR spectra of CIP/EPDM composite absorbing ▶ material with different phr of antioxidant RD before and after hot air aging

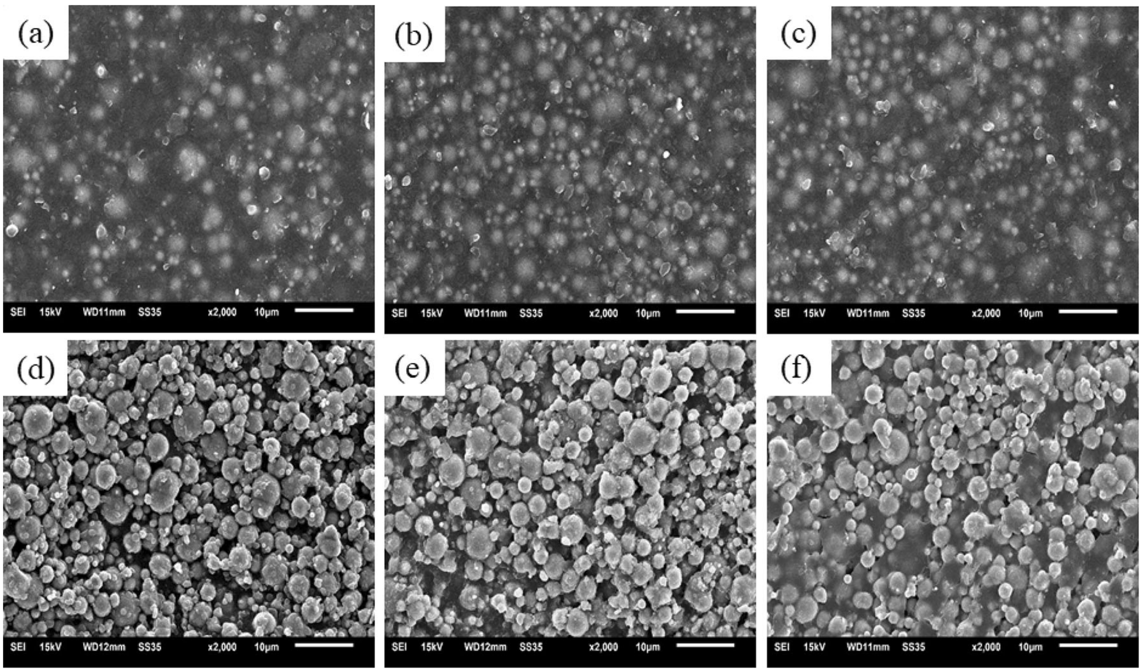


Fig. 5 SEM images of the surface of CIP/EPDM composite absorbing material with 1.0 phr of antioxidant RD before and after hot air aging: **a** 0 d, **b** 1 d, **c** 4 d, **d** 8 d, **e** 12 d, and **f** 14 d

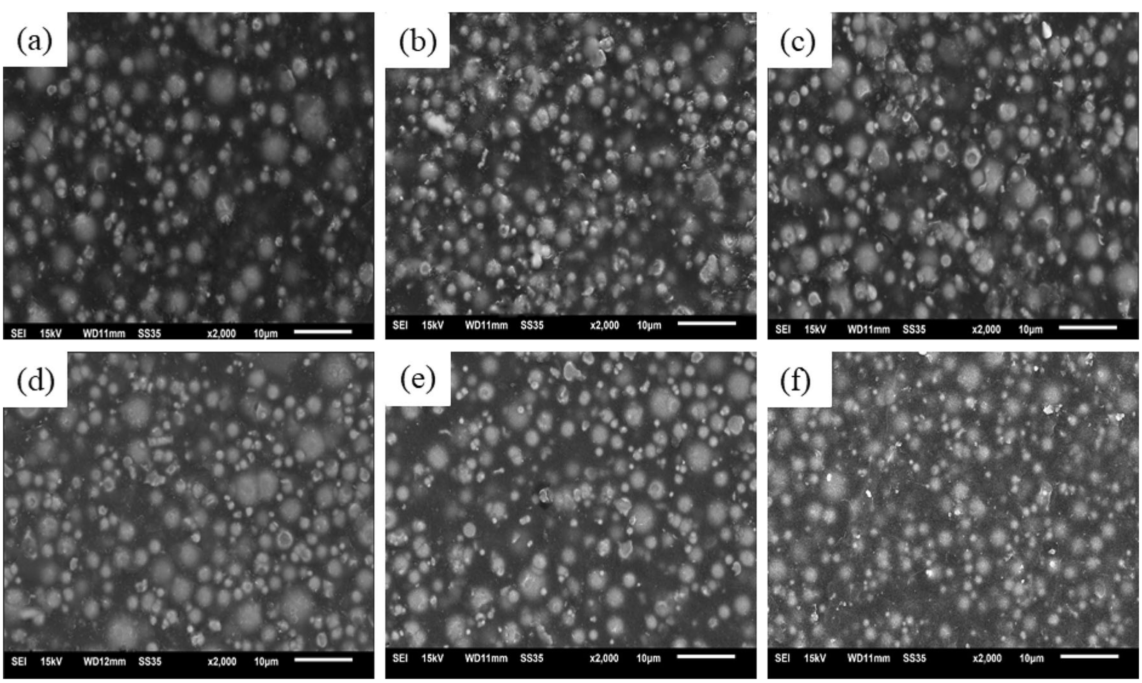
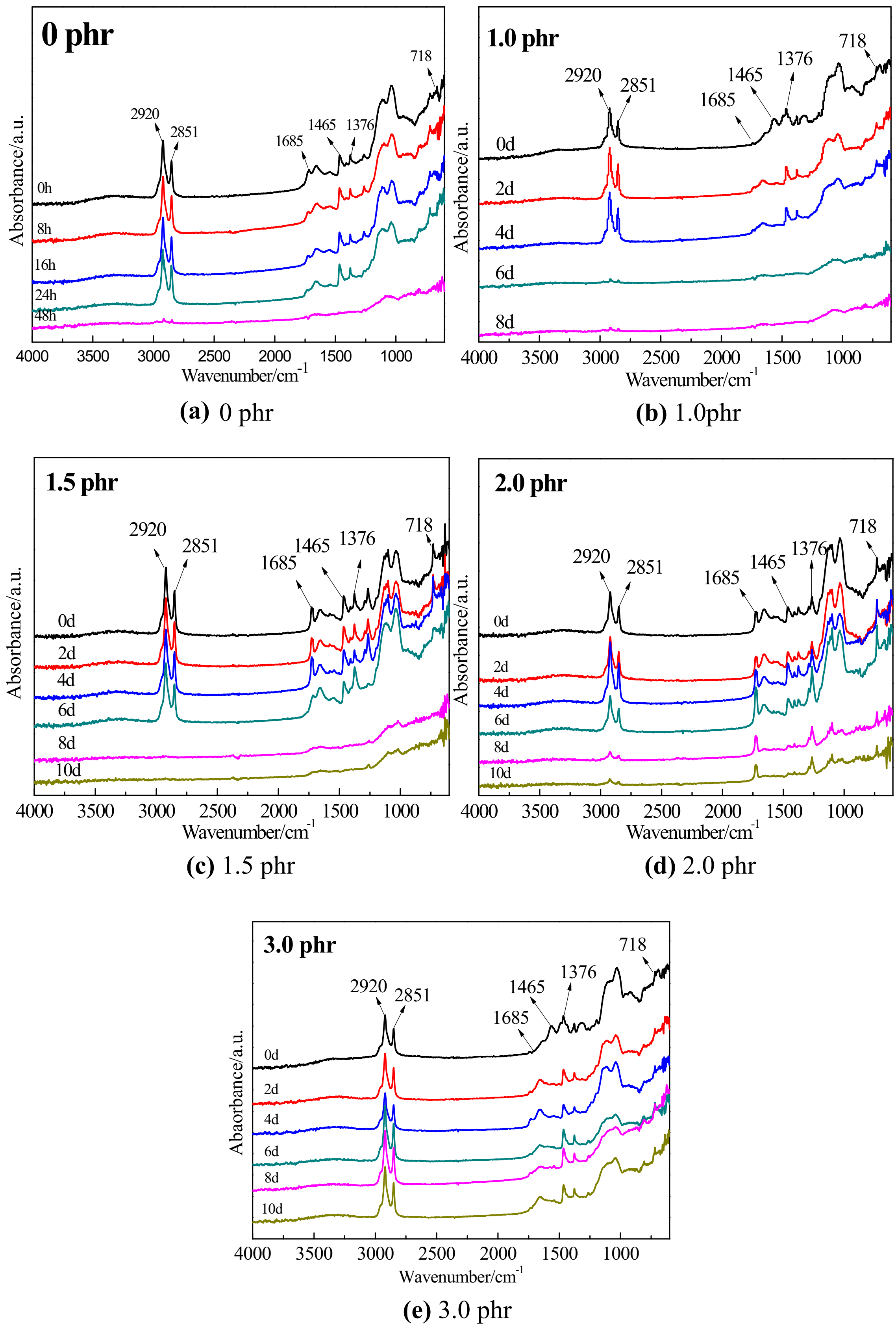


Fig. 6 SEM images of the surface of CIP/EPDM composite absorbing material with 3.0 phr of antioxidant RD before and after hot air aging: **a** 0 d, **b** 1 d, **c** 4 d, **d** 8 d, **e** 12 d, and **f** 14 d



(a) 0 phr **(b)** 1.0phr

(c) 1.5 phr **(d)** 2.0 phr

(e) 3.0 phr

RD content, the time of the attenuation or disappearance of the infrared absorption peak of the composite absorbing material is delayed.With RD content of 0 phr, the aging after 2 days disappear, and with RD content of 1.0 phr, the aging after 4 days disappear. When RD content was 1.5 phr, aging after 6 days disappear, while when RD content increased to 2.0 phr, although aged 8 days, intensity of infrared characteristic peaks decreased significantly, but was not completely disappeared, and when RD content was 3.0 phr, 14 days after the infrared characteristic peaks of aging still keep good strength, with no signs of decay. The above results were consistent with the results of scanning electron microscopy. The addition of RD delayed the aging of the composite absorbing materials and slowed the rate of surface morphology change. Moreover, the more the amount of RD was added, the more obvious the effect was. The rate of small molecule escape on the surface of EPDM was slowed down due to the oxidation cross-linking, that is, under the action of antioxidant RD, the rate of decrease of EPDM content on the surface was slowed down, so the attenuation of intensity of infrared characteristic peak on the surface was correspondingly slowed down.

In summary, the antioxidant RD has a significant effect on delaying the aging rate of samples, and the effect is more obvious with the increase of RD content. At the same time, due to the addition of more than 3.0 phr, vulcanized rubber will appear with spray frost phenomenon and so the best addition of 3.0 phr.

3.4 The TG of CIP/EPDM composite absorbing material with different dosage of antioxidant RD after the same time of hot air aging

In order to verify the inhibitory effect of antioxidant RD on the cross-linking of EPDM catalyzed by oxygen and CIP, TG analysis was performed on samples of different antioxidant RD. Figure 8 shows the TG curve of CIP/EPDM composite materials with different RD loads after 7 days of hot air aging. It can be seen from the figure that with the increase of RD content, the maximum weight loss of composite absorbing materials increases gradually at the same aging time. This is because RD can effectively inhibit the cross-linking degree between CIP and EPDM under aerobic conditions, and the higher the RD content, the more obvious the effect. The cross-linking degree of the composite absorbing material with less RD is larger after hot air aging, so the thermal stability is better and the maximum weight loss is smaller after aging.

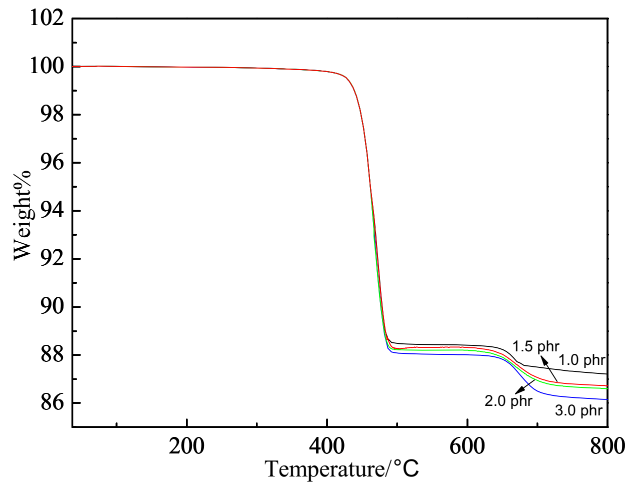


Fig. 8 TG curves of CIP/EPDM composite absorbing material with different phr of antioxidant RD after hot air aging for 7 d

3.5 Effects of different RD contents on the electromagnetic properties of CIP/EPDM composite absorbing materials before and after hot air aging

Figures 9 and 10 show the electromagnetic performance changes of CIP/EPDM composite materials with 0 phr and 3.0 phr antioxidant RD at different aging time.It can be seen from Fig. 9 that the addition of RD did not change rule of the dielectric constant, but compared with the composites without antioxidant, when RD content was 3.0 phr, the real and imaginary parts of the complex dielectric constant fluctuated very little.

Figure 10 shows the change of complex permeability of CIP/EPDM composite material with different parts of antioxidant RD before and after hot air aging. It can be seen from the figure that the real part of the composite permeability of the composite material still has no obvious change, and the imaginary part of the composite permeability has a smaller change with the aging time.

Figure 11 shows the change of reflectivity of CIP/EPDM composite with different antioxidant RD content before and after hot air aging. As can be seen from the figure, when the antioxidant RD was 0 phr,

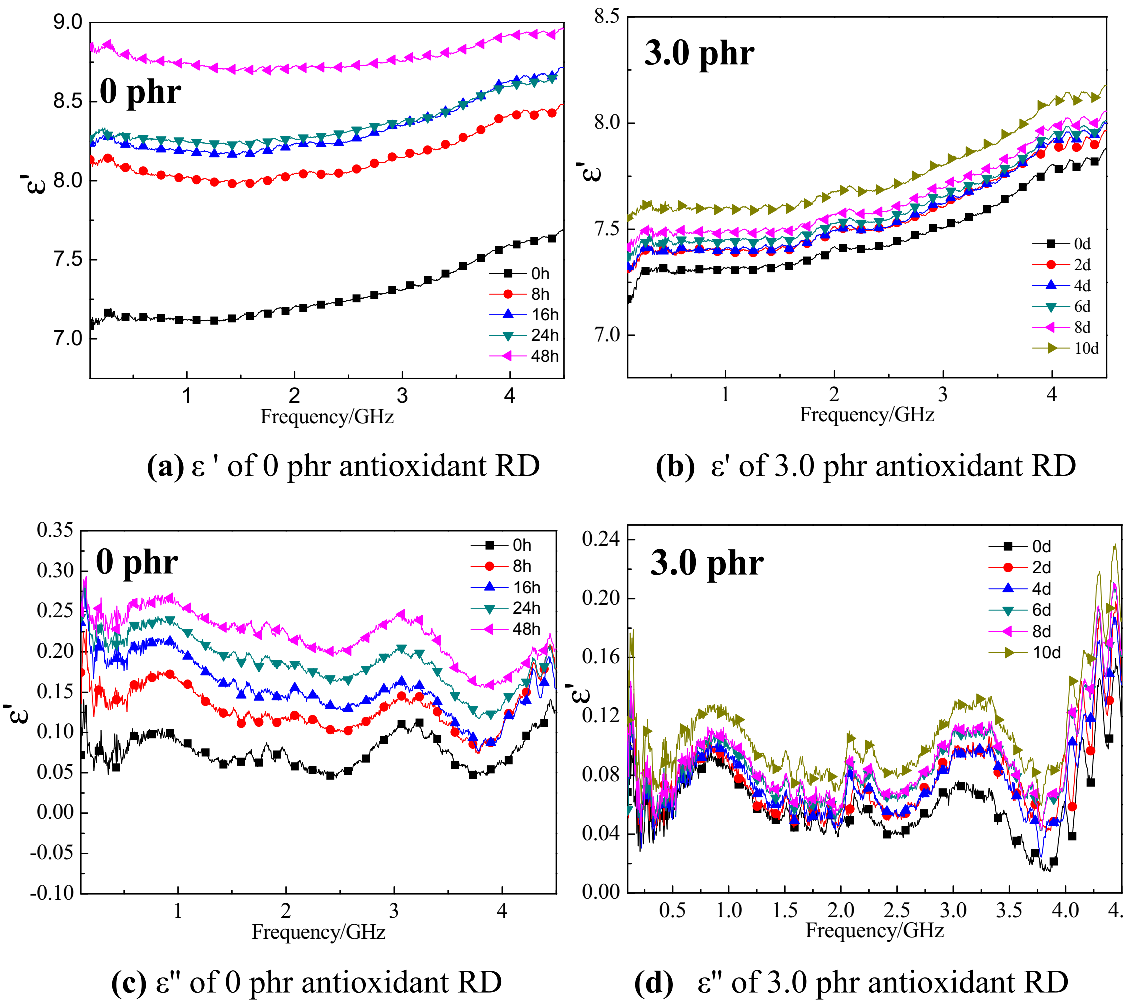


Fig. 9 The ε′ and ε″ of CIP/EPDM composite absorbing material with different phr of antioxidant RD before and after hot air aging

the minimum reflectance changed from − 22.16 to − 17.3941dB, the minimum reflection frequency fluctuated between 3.64GHz and 4.12GHz, and the effective absorption bandwidth (RL < − 10dB) also decreased from 2.23 to 2.04GHz.When the antioxidant RD was 3.0 phr, the minimum reflectance changed from − 19.91 to − 16.81dB with the increase of the aging time of hot air, the minimum reflectance fluctuated between 4.00GHz and 4.19GHz, and the effective absorption bandwidth (RL < − 10dB) also decreased from 2.25 to 2.06GHz.It can be found that the fluctuation of the lowest reflectance, the lowest reflection frequency, and the effective absorption bandwidth of the composite material decreases after adding antioxidant RD, and the reflectance curves before and after hot air aging almost completely overlap.

According to the above results, the change rate of the electromagnetic properties of CIP/EPDM composite materials was reduced by the addition of antioxidant RD, which indicated that the antioxidant RD could effectively delay the aging of the composite absorbing materials and make them maintain good mechanical and electromagnetic properties after a long time of hot air aging.

In general, through the above analysis, the addition of antioxidant RD can effectively alleviate the aging problem of CIP/EPDM composite materials caused by high temperature and increase its service life from 2 days to more than 10 days. It has certain reference value for the production and preparation of high-temperature resistant rubber composite wave absorbing materials.

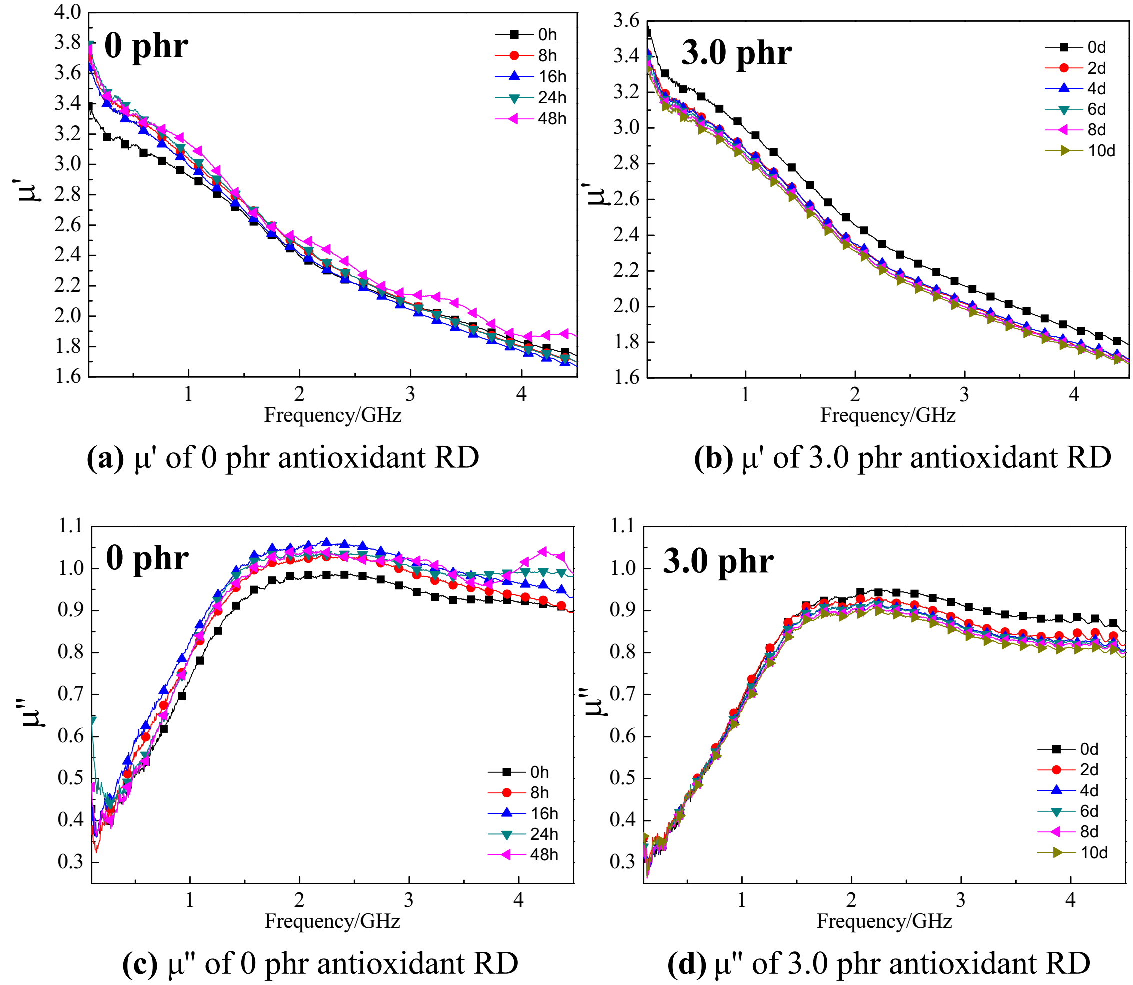


Fig. 10 The μ′ and μ″ of CIP/EPDM composite absorbing material with different phr of antioxidant RD before and after hot air aging

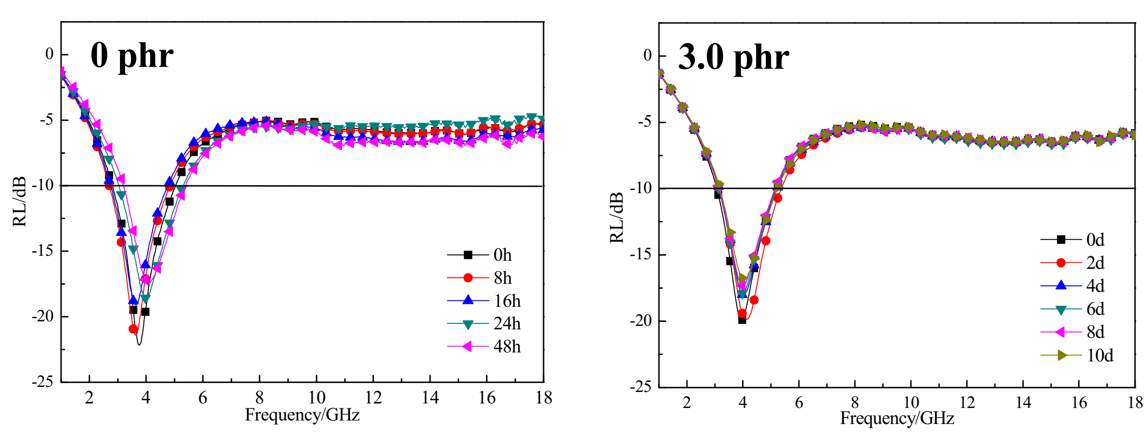


Fig. 11 The RL of CIP/EPDM composite absorbing material with different phr of antioxidant RD before and after hot air aging

4 Conclusion

In this research, the effect of the loading of antioxidant RD on the CIP/ EPDM composite absorbing material under high-temperature environment has been studied. Through mechanical properties, SEM, ATR-FTIR, and TG analysis, it is found that the composite material loaded with 0 phr antioxidant RD will fail due to the aging of the rubber matrix at 150 °C for 2 days. With the increase of load, the service life of the composite gradually increases. When the load is 3 phr, the service life exceeds 10 d. However, due to frost spraying, antioxidant RD load cannot be higher than 3 phr.In the electromagnetic performance test, the minimum reflectance of the sample with the antioxidant RD load of 0 phr increased from − 22.16 to − 17.39dB within two days, and the effective absorption bandwidth (RL < − 10dB) was reduced from 2.23 to 2.04 GHz with big fluctuations. And the minimum reflectance of the sample with the antioxidant RD loading of 3 phr increased from − 19.91 to − 16.81dB within ten days, and the effective absorption bandwidth was reduced from 2.25 to 2.06 GHz with small fluctuations. Therefore, loading antioxidant RD can effectively delay the aging rate of CIP/EPDM composite absorbing materials.

In general, loading 3 phr antioxidant RD can significantly delay the aging rate of CIP/EPDM composite absorbing materials. Antioxidant RD has shown good application prospects in resin-based composite absorbing materials that use magnetic materials as absorbents.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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